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NRL Report 5692

FISSION PRODUCT RADIOACTIVITY IN THE AIR ALONG THE 80TH MERIDIAN (WEST) DURING 1960

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ABSTRACT

Since 1958, the U. S. NRL has had a continuing study of radioactivity in air along the 80th meridian (West). The year 1960 brought progressive decreases in airborne fission product radioactivity though the rate of decrease with time was significantly less than during 1959. During 1960 the activity levels in the Northern and Southern Hemispheres gave indications of approaching equilibrium which suggested that transequatorial mixing in the stratosphere does occur but with a mean lifetime of several years for the process. However, two years after the cessation of the major nuclear test programs, differences in the fission product compositions of the two hemispheres still exist.

The relatively small amounts of radioactivity generated by the French nuclear tests in the Sahara produced only transient effects at a few sites along the 80th meridian. The interpretation of atmospheric mixing patterns from measurements of the concentrations of the longer-lived fission products in the air was not significantly affected by these fresher debris. Fission product ratios involving some of the shorter-lived fission products did document the presence of debris from the February 1960 test in a band extending from Miami, Florida, to Guayaquil, Ecuador, with the suggestion that small amounts of debris did appear in other areas.

PROBLEM STATUS

This is an interim report. Work on this problem is continuing.

AUTHORIZATION

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FISSION PRODUCT RADIOACTIVITY IN THE AIR ALONG THE 80TH MERIDIAN (WEST) DURING 1960

INTRODUCTION

The program of measurement of the fission product radioactivity in the air at ground level along the 80th meridian during the past years has proved exceedingly worthwhile in correlating the latitudinal variations of radioactivity with tropospheric and stratospheric sources of nuclear debris (1-3). The absence of any large scale nuclear testing during the past two years has permitted the investigation of stratospheric deposition processes with a minimum of interference from similar debris introduced directly into the troposphere. In 1960, radioactivity levels, except for the period immediately following the French nuclear tests in the Sahara (4), were sufficiently low to require some modification of previously used collection schemes to maintain the desired statistical accuracy in counting. Further changes in the future are indicated as the fission product concentration continues to decrease. The low activity levels, unfortunately, required a decrease in the number of collecting stations in operation during 1960 because of the long counting times required of each sample to obtain reasonable statistical accuracy. The cooperating sites during 1960 are listed in Table 1; the choice was made on the basis of geographical location, with emphasis on sites at low altitudes.

This program, as in past years, has been operated by the U. S. Naval Research Laboratory with the cooperation of interested agencies of the United States, Canada, Ecuador, Peru, Bolivia, and Chile which have made the actual sample collections and forwarded them to NRL for analysis. Partial financial support of this program has been obtained from the Division of Biology and Medicine, U. S. Atomic Energy Commission.

EXPERIMENTAL PROCEDURE

Continuous sampling of particulate radioactive material in the air at ground level was performed at 11 low altitude sites along the 80th meridian (west) and at two high altitude sites, Chacaltaya, Bolivia, and Mauna Loa, Hawaii, at comparable latitudes north and south of the equator.

The sampling procedure involved drawing air continuously at a known rate (approximately 1200 cubic meters per day) through high efficiency filters 8 inches in diameter by use of positive displacement blowers. Filters were changed three times per week and forwarded immediately to NRL for assay for gross β activity two weeks after collection, as described elsewhere (2). Radiochemical analyses were performed on collections from each site for the months of January, March, May, July, September, and November 1960.

The longer sampling times were necessitated by the lower fission product activity prevalent in the air during 1960. This lower activity also required the use of low-level counting techniques (anticoincidence counting in heavily shielded equipment) to obtain the desired accuracy in measurement of the radiochemically separated nuclides. These counters were standardized in late 1959 and again in early 1961 using radioactive standards of known disintegration rates.

Sr^{90} from the various samples was determined by separating out and counting the Y^{90} daughter activity and also by counting the equilibrium mixture of Sr^{90}-Y^{90} and comparing

Table 1
Collecting Sites Associated With the NRL 80th Meridian
Air Sampling Program During 1960

Station	Latitude	Longitude	Elevation (m)	Operator
Thule, Greenland	76 35'N	68 35'W	250	Geophysics Research Directorate, USAF, Cambridge Research Laboratories
Moosonee, Ontario, Canada	51 16'N	80 39'W	10	Meteorological Branch, Department of Transport (Canada)
Washington, D. C.*	38 50'N	76 57'W	88	U. S. Weather Bureau
Miami, Florida	25 49'N	80 17'W	4	U. S. Weather Bureau
Mauna Loa, Hawaii	19 28'N	155 36'W	3394	U. S. Weather Bureau
San Juan, Puerto Rico†	18 26'N	66 00'W	10	U. S. Weather Bureau
Miraflores, Panama Canal Zone	9 00'N	79 35'W	10	Canal Zone Corrosion Laboratory (U. S. Naval Research Laboratory)
Guayaquil, Ecuador	2 10'S	79 52'W	7	Meteorological Office, Direccion General de Aviacion Civil
Lima, Peru	12 06 S	77 01'W	134	Corporacion Peruana de Aeropuertos y Aviacion Comercial (CORPAC)
Chacaltaya, Bolivia	17 10'S	68 15'W	5220	Universidad Mayor de San Andres, Laboratorio de Fisica Cosmica de Chacaltaya
Antofagasta, Chile	23 37'S	70 16'W	519	NASA Satellite Tracking Station
Santiago, Chile	33 27'S	70 42'W	520	Oficina Meteorologica de Chile
Puerto Montt, Chile	41 27'S	72 57'W	5	Oficina Meteorologica de Chile
Punta Arenas, Chile	53 08'S	70 53'W	3	Oficina Meteorologica de Chile

*Collecting equipment moved from Silver Hill, Maryland, to Sterling, Virginia (38 54'N, 77 26'W, elevation 82 m) in September 1960.
†Samples collected routinely analyzed for gross β activity only during periods of nuclear testing by the USSR.

it with a similarly prepared Sr^{90}-Y^{90} standard source. In the absence of fresh debris containing Sr^{90}, as determined by the absence of radioactivity in the Ce^{144} and Y^{91} fractions, the latter procedure is preferred because of its inherently greater accuracy due to the fewer processing steps required.

RESULTS

Gross Fission Product Concentrations in the Air

The monthly average gross fission product concentrations are listed in Table 2. Monthly profiles of the gross fission product β activity in the ground-level air along the 80th meridian are shown in Fig. 1. There are several points of interest in these profiles. First, the great, but short-lived, influence of the French test of February 13, 1960 on the gross radioactivity in the region between 26 N and 2 S is apparent. The absence of debris from this test in other areas was confirmed by radiochemical analyses which indicated

Table 2
Gross Fission Products in the Ground-Level Air During 1980

Site	Activity (dis/min per m³ of air)											
	Jan.	Feb.	Mar.	Apr.	May	June	July	Aug.	Sept.	Oct.	Nov.	Dec.
Thule	0.29	0.36	0.36	0.35	0.22	0.21	0.18	0.15	0.07	0.05	0.06	0.09
Moosonee	0.20	0.34	0.42	0.41	0.29	0.38	0.27	0.16	0.12	0.09	0.08	0.13
Washington	0.25	0.35	0.43	0.53	0.49	0.48	0.34	0.24	0.18	0.17	0.14	0.15
Miami	0.42	0.57	0.97	0.55	0.53	0.24	0.13	0.14	0.07	0.09	0.14	0.18
Miraflores	0.17	3.65	5.55	0.22	0.06	0.03	0.05	0.05	0.03	0.03	0.03	0.05
Guayaquil	0.06	0.08	0.47	0.07	0.07	0.05	0.05	0.05	0.04	0.03	0.06	0.05
Lima	0.04	0.04	0.05	0.04	0.02	0.03	0.04	0.03	0.06	0.06	0.05	0.05
Antofagasta	0.08	0.10	0.12	0.08	0.07	0.07	0.09	0.09	0.13	0.13	0.10	0.10
Santiago	0.11	0.15	0.13	0.09	0.08	0.07	0.08	0.09	0.12	0.09	0.10	0.13
Puerto Montt	0.09	0.10	0.10	0.05	0.05	0.03	0.02	0.03	0.05	0.04	-	-
Punta Arenas	0.03	0.04	0.04	0.03	0.05	-	0.02	0.02	0.03	0.02	0.02	0.05
Mauna Loa	0.24	0.34	0.82	0.27	0.23	0.24	0.23	0.12	0.19	0.07	0.07	0.05
Charaltaya	0.02	0.04	0.06	0.04	0.05	0.04	0.05	0.06	0.05	0.06	0.05	0.06

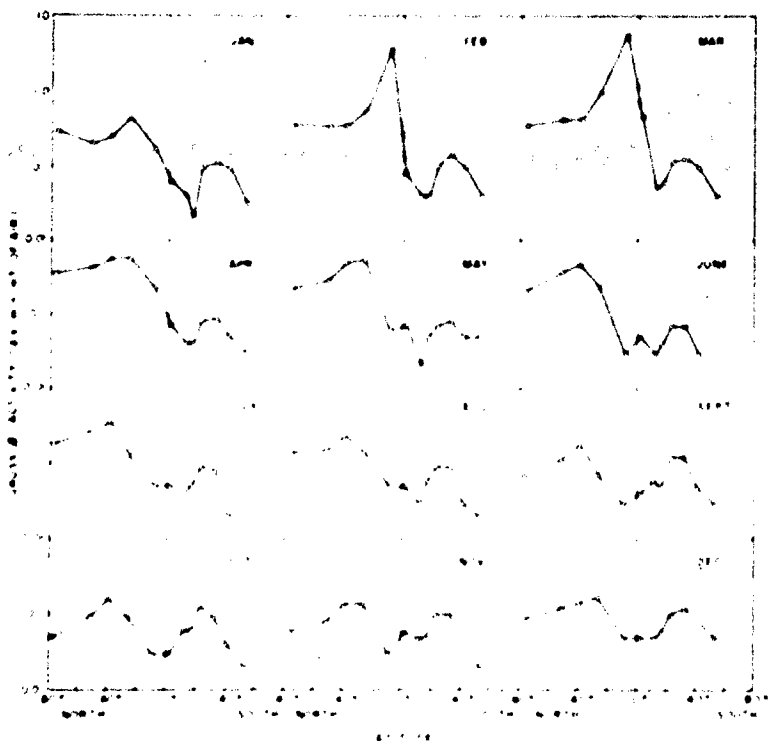


Fig. 1 – Profiles of the gross fission product concentrations in the ground-level air along the Atlantic coast of the Americas

negligible quantities of some of the shorter-lived fission products (Ce^{141}, Sr^{89}, Y^{91}, Pr^{143}, Nd^{147}, etc.) in collections from these areas. A more detailed presentation of radioactivity changes associated with this first French atomic test is given in Fig. 2. The effect of the passage of the radioactive cloud or clouds would be even more apparent (particularly at Miami) if the results had been plotted on a linear scale, rather than on a logarithmic scale. Due to the background activity remaining after the first French test and the increasing rate of stratospheric "fallout" during this season, debris from the second smaller French test could not be identified. Some information on these nuclear tests is included in Table 3. Analyses of collections made in May 1960 indicated that removal of debris from these tests was essentially complete during the period of 1 to 2-1/2 months which had elapsed since the explosions. This rapid deposition of material introduced below the tropopause is to be expected and, in fact, may be even more rapid in these latitudes than elsewhere due to the frequent occurrence of heavy rainfall in the tropics.

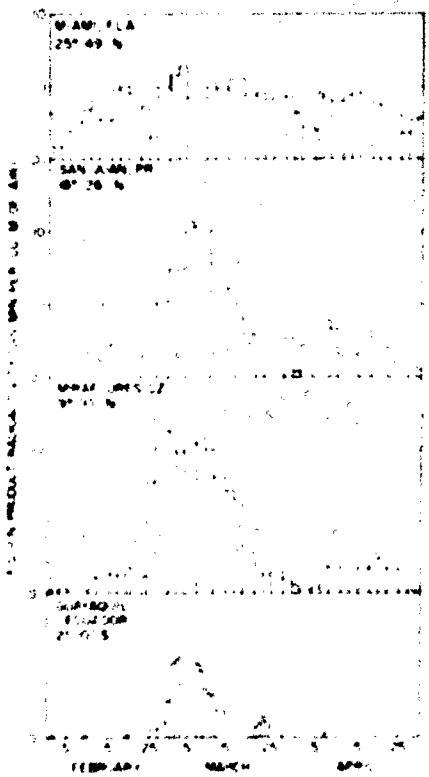


Fig. 2 - Changes in the concentration of gross fission product radioactivity in the air along the 80th meridian (West) following the French nuclear tests of 1960

During 1960 the gross activity in the ground-level air of the Northern Hemisphere underwent the expected seasonal changes, though they were confused to some extent by the French tests. These seasonal changes are more apparent in the Sr^{90} data presented in a later section. It is of interest that in the Southern Hemisphere, while there were less obvious seasonal effects, the overall gross fission product concentrations did not decline markedly during the year. This would indicate a replenishment of the Southern Hemisphere stratospheric source at a rate commensurate with that by which it was being depleted by deposition and radioactive decay.

Table 3
Information on the 1960 French Nuclear Tests
in the Sahara Desert Near Reggane, Algeria

Date	Yield*	First Arrival of Debris at 80th Meridian
February 13	60 kt	Between February 24-26, 1960 (see Fig. 2)
April 1	5 kt	Not identified
December 27	5 kt	Between January 8-9, 1961

*Unofficial – information obtained from newspaper accounts.

Progressive changes in the gross fission products in the air at several corresponding Northern and Southern Hemisphere sites during the past two years are shown in Fig. 3. During succeeding seasonal minima in the Northern Hemisphere, the air concentrations of fission products at Washington and Miami dipped lower and lower until, in the fall of 1960, the activity levels at these sites were no higher than at Antofagasta and Santiago. It might be expected that at the seasonal minimum during 1961, the Northern Hemisphere values will lie much below those of the Southern Hemisphere. With depletion of the excess radioactivity in the northern stratosphere, the corresponding seasonal effects in the Southern Hemisphere should also become more evident since the relative quantities of debris crossing from North of the equator would be less significant.

The radioactivity patterns during 1960 exhibited the same form as has been conspicuous during past years, namely, maxima in the midlatitudes of each hemisphere with a minimum in the Tropics. The highest levels of activity were at Miami, Florida, during the winter and spring of 1960, shifting northward to Washington, D. C., for the summer and autumn seasons, in a manner observed in prior years (2). A similar, though weaker, northward shift appeared to occur at the same time in the Southern Hemisphere.

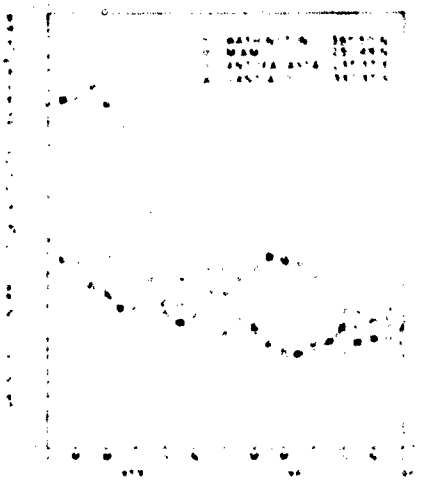


Fig. 3 – Progressive changes in the air concentration of gross fission products at corresponding sites in the Northern and Southern Hemispheres

A further item of note is the consistently large difference in radioactivity concentrations at Moosonee (51 N) as compared to Punta Arenas (53 S), which exceeds greatly the ratios of activity at any other corresponding sites. This may be perhaps attributable to the lack of any significant source of radioactivity in the lower antarctic stratosphere or perhaps to a difference in the meteorology of the two regions. Other investigators have reported the influx of fission products and of Rh^{102} tracer from high altitude shots of the Hardtack series into the upper stratosphere at the higher latitudes of both hemispheres (5); the information presented in this report suggests that radioactive debris from these tests (Teak, Orange) have as yet had no measurable effect on the distribution of radioactivity at the earth's surface.

Radiochemical Analyses

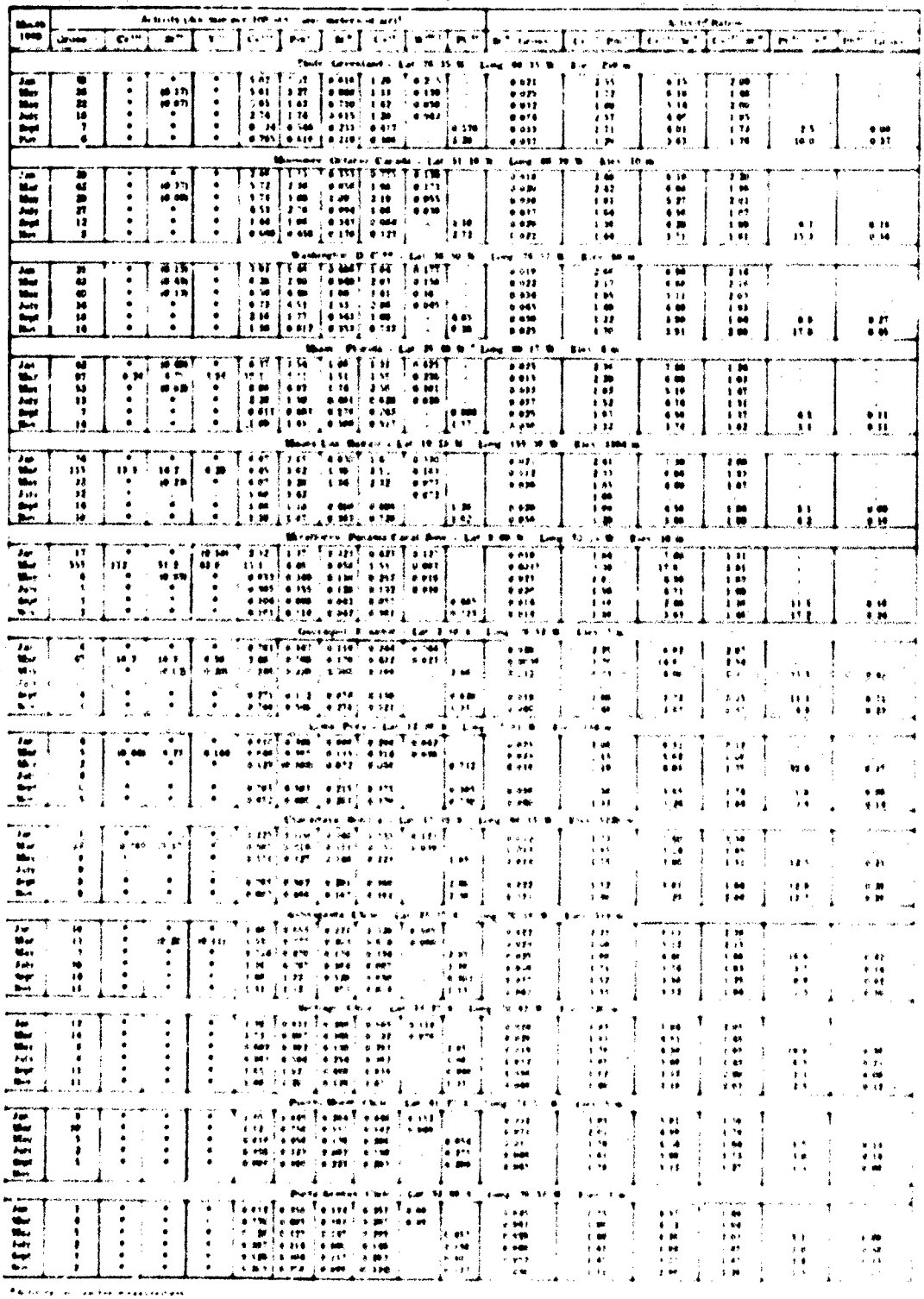
A summary of the radiochemical analyses of composite monthly air-filter collections made during 1960 is presented in Table 4 in units of disintegrations per minute per 100 standard cubic meters of air and corrected for radioactive decay to the midpoint of the collection period. Several activity ratios are also listed; these are discussed in later sections of the text. Where available, preliminary data from the radiochemical analyses of combined January-February 1961 samples are included in the graphical presentations which follow.

Sr^{90} in the Air

The average Sr^{90} concentrations in the air at the various sites during alternate months are shown graphically in Figs. 4-6. In the North Temperate Zone (Fig. 4), the spring maximum of 1960 is well defined, as was the 1959 maximum, with a peak (spring 1960) to valley (winter 1960-61) ratio varying from about 3 at Washington to 10 at Miami. This may be compared to 1959 with maximum/minimum ratios in the range of 19 to 42. The effective half-time of decrease of the Sr^{90} concentration following the spring peak was about 40 days during the interval July-September 1960 compared with 30 days during 1959. The changes are attributable to depletion during 1959 of much of the excess stratospheric radioactivity associated with the USSR tests of 1958. Interestingly, the maxima and minima appeared earliest at the Miami site during both 1959 and 1960; during 1960 these changes occurred earlier at Thule than either at Moosonee or Washington.

In the South Temperate Zone (Fig. 5), Sr^{90} concentrations show definite increases at Antofagasta and Santiago during the Southern Hemisphere spring season. However, this seasonal effect is not as pronounced at Puerto Montt and Punta Arenas and occurs at a later time. In the Tropics (Fig. 6) there is a rather distinct out-of-phase relationship in the Sr^{90} concentrations at the sites above and below the equator. In addition to the seasonal maxima and minima there may be other effects associated with changing rainfall patterns (dry and rainy seasons) which can distort the general activity levels. The French nuclear test of February 1960 also contributed to the Sr^{90} activity during March, most importantly at Miraflores and Guayaquil. There is an unexplainable secondary peak in atmospheric radioactivity at all Southern Hemisphere sites during March which does not appear to be related either to the French tests or to activity levels in the Northern Hemisphere.

Profiles of Sr^{90} along the 80th meridian during the periods of the seasonal maxima and minima are shown in Fig. 7. For comparison the profile for January 1960 is shown to indicate the distribution during an intermediate period. It should be noted that the data for no one month will depict accurately the profiles during periods of maxima and minima because of the different times of occurrence at the various sites. It is evident, however, that decided seasonal changes do occur which are roughly six months out-of-phase, as might be expected. The position of minimum activity always lies within the tropics but



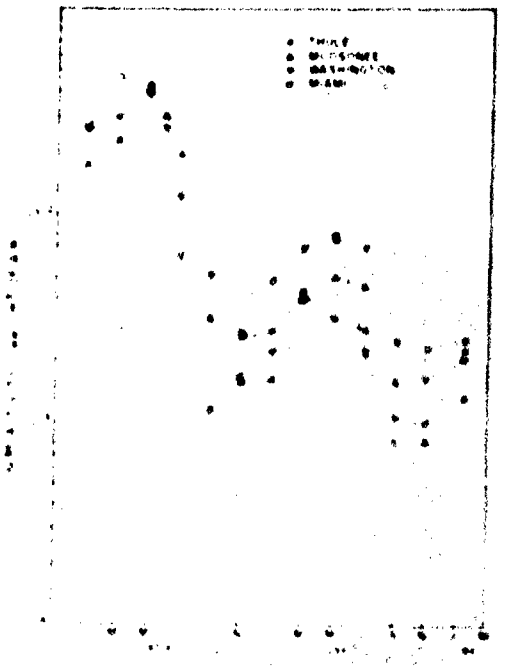


Fig. 4 - Concentrations of Sr^{90} in the air of the North Temperate Zone [1958-60]

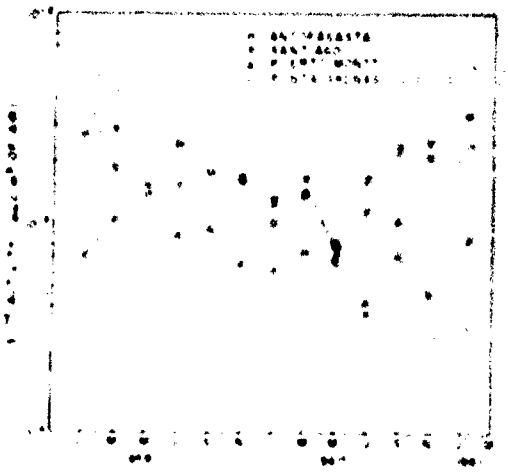


Fig. 5 - Concentrations of Sr^{90} in the air of the North Temperate Zone [1958-60]

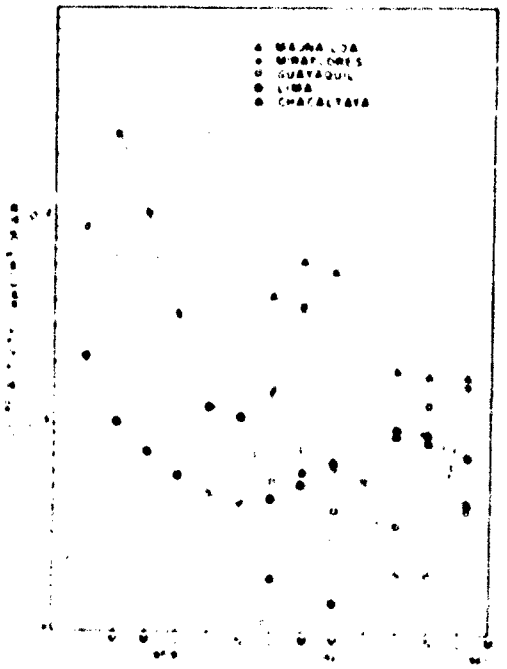


Fig. 6 – Concentration of Sr^{90} in the air of the Tropic Zone [illegible]

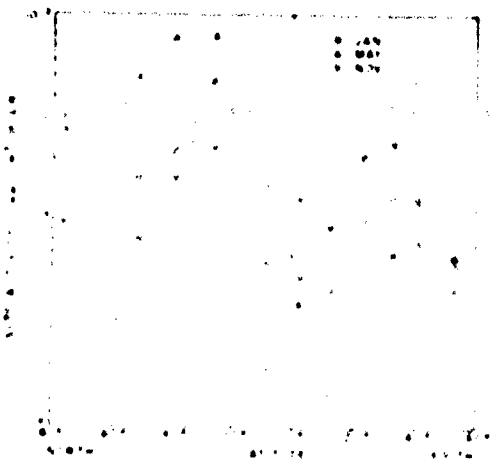


Fig. 7 – Deposition of Sr^{90} and rainfall along the [illegible] during [illegible], May, and September 1960

shifts with time toward the area of the seasonal minimum. This change may be due to the shift of a belt of rainfall with season associated with changes in the meteorological equator.

Contribution of Sr^{90} to the Gross Fission Product Activity in the Air

In past years an indication of an apparent age of radioactive debris being collected could be estimated from the ratio of Sr^{90} to the gross β activity (3). During 1960 the low levels of gross fission product activity have made its estimation of less reliability; more significantly, however, the relative contribution of the long-lived natural radioactivity (RaD-RaE) has increased to a point where it may be contributing a sizeable fraction of the total β activity measured (Table 4). Because of the incompleteness of data on this radioactive component, it is not practical to make a correction for its contribution to the gross β activity. It is evident, on the other hand, that fresh activity with its characteristic low Sr^{90}/gross β activity ratio was collected at Miraflores, Mauna Loa, Miami, and Guayaquil during March 1960.

Activity Ratios

Ce^{144}/Sr^{90} – The ratios of Ce^{144} activity to Sr^{90} activity in the air at corresponding areas of the Northern and Southern Hemispheres during 1959 and 1960 are shown in Fig. 8. Following the spring peak of 1959 in the Northern Hemisphere the Ce^{144}/Sr^{90} ratio decreased more rapidly than the rate expected for radioactive decay alone. This effect was the result of the depletion of the younger stratospheric source of debris from the USSR October 1958 test series with the resulting relatively greater contribution of older debris from more southerly located stratospheric sources. During the spring of 1960 there was no indication of a second influx of younger debris from the USSR source, confirming its essential depletion or complete integration through mixing during the preceding year. In the Southern Hemisphere there appeared to be some influx of older debris during September-November 1959 which is consistent with the increased contribution of older debris from an antarctic source (resulting from the South Poleward migration of tropical debris). No such change was evident during 1960.

The changes in the Ce^{144}/Sr^{90} activity ratios during 1960 do indicate a slowly approaching equilibrium as limited trans-equatorial mixing progresses. A comparison of the Ce^{144}/Sr^{90} activity ratios in the two areas during July 1959 with those during July 1960 indicates a lessening of the age differential between the fission product conglomerates in the stratospheric sources of supply; this age difference decreased from about 7 months to about 2 months in a year's time.

Ce^{144}/Pm^{147} – The decrease in the Ce^{144}/Pm^{147} activity ratios with time is shown in Fig. 9 and is essentially at the rate expected. The difficulty in mounting the Pm^{147} sample with its β particle of low energy so as to reproduce reliably the self-absorption characteristics of the sample is probably the major cause for the spread in values of this ratio. There appear to be no systematic differences in the Ce^{144}/Pm^{147} ratios in the samples from the two hemispheres in contrast to the Ce^{144}/Sr^{90} ratios.

Cs^{137}/Sr^{90} – The Cs^{137}/Sr^{90} activity ratios listed in Table 4 are generally in the neighborhood of 1.9 (50% in range 1.90 ± 0.15) with a secondary grouping below 1.5. All of the Miami samples had ratios in this latter grouping; the significance of this difference is unknown.

The variation in counting rate of cesium samples has been found in many cases to be due to the size of the crystallites of $CsClO_4$, which is the mounting form of the carrier. The larger aggregates can cause an appreciable loss in the measured activity due to the increased self-absorption of the sample. However, this factor does not account for the fact that all of the Miami samples have low Cs^{137}/Sr^{90} activity ratios.

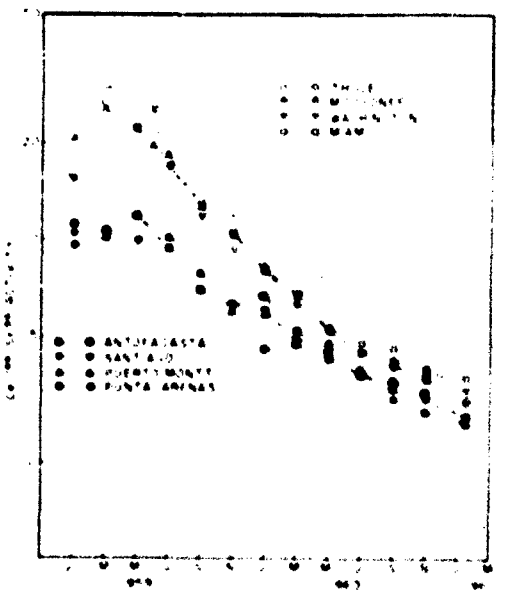


Fig. 8 - Ratios of Ce^{144} to Sr^{90} activity at various sites during 1959-60

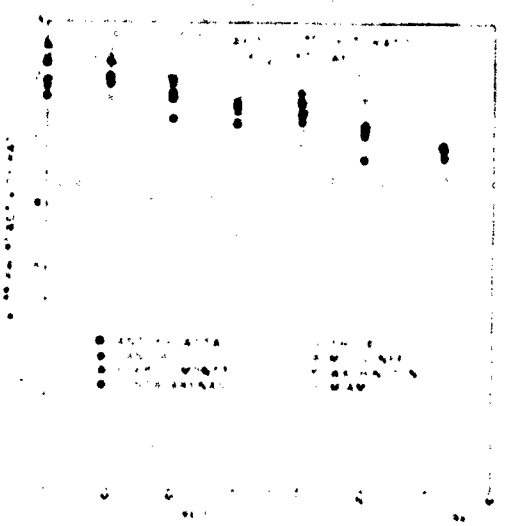


Fig. 9 - Ratios of Ce^{144} to Ba^{137} activity at various sites during [illegible]

Effect of Altitude on Relative Fission Product Concentrations

The changes in the relative activity levels at a pair of high altitude sites and also at a pair of sites near sea level at comparable latitudes North and South of the equator are shown in Fig. 10. The low altitude sites show the accentuated peak-to-valley relationship expected as a result of out-of-phase seasonal changes in deposition rates in the two hemispheres. This same relationship is found for gross β, Pm^{147}, and Cs^{137} as well as for the depicted Sr^{90} and Ce^{141} activities. An entirely different pattern, confirmed by all the activities measured, is evident in the comparison of activity concentrations at the high altitude sites. The high value of the north-to-south ratio in January 1960 is due to the low activity collected at Chacaltaya at that time; the 1961 gross β activity measurements indicate that a more normal variation will be obtained during 1961. Perhaps the only significance of these data is that it indicates that the activities at ground level are not necessarily related to those at higher levels overhead in the troposphere. Peirson, et al., have already pointed out a difference between stratospheric and tropospheric air over England (6).

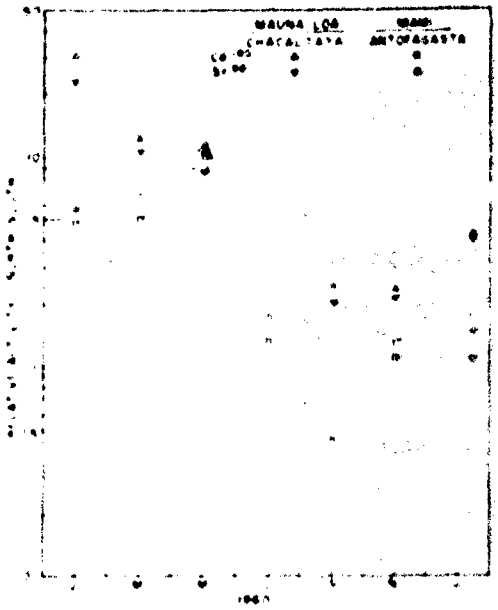


Fig. 10 – Effect of altitude on the relative fission product concentrations at selected stations north and south of the equator

Contributions of Hardtack Debris to Atmospheric Contamination During 1960

As explained in detail in previous reports (1-3), the W^{185} produced uniquely in the U S Hardtack series at the Pacific Proving Grounds during 1958 can be used to estimate the contribution of these tests to the general atmospheric contamination. There was sufficient W^{185} activity remaining in the air during 1960 for reasonable determinations of this isotope to be made only through March in the Southern Hemisphere and through July in the

Northern Hemisphere. Removal processes as well as radioactive decay were responsible for the low levels of W^{185} activity encountered.

The contribution of Sr^{90} from the U.S. Hardtack tests to the total Sr^{90} in the air at various sites during early 1960 is shown in Table 5. These calculations are based on the assumption of a W^{185}/Sr^{90} activity ratio of 500 for Hardtack debris as of July 15, 1958. The contributions of Hardtack Sr^{90} to the total were essentially unchanged (within experimental error) when compared with corresponding periods of 1959 (3). In both 1959 and 1960 the percentage of Hardtack debris decreased markedly during the spring in the Northern Hemisphere; during the periods January-March of 1959 and 1960 no significant changes occurred in the Southern Hemisphere.

Table 5
Contribution of Sr^{90} From U.S. Hardtack Tests to the Total Sr^{90} in the Air During 1960

Site	Hardtack Sr^{90} (percent of total Sr^{90})			
	Jan.	Mar.	May	July
Thule	14	9	8	13
Moosonee	13	12	5	7
Washington	12	10	6	6
Miami	14	9	6	11
Mauna Loa	12	7	7	-
Miraflores	14	6	15	16
Guayaquil	13	8	-	-
Lima	14	16	-	-
Chacaltaya	11	18	-	-
Antofagasta	16	12	-	-
Santiago	14	13	-	-
Puerto Montt	18	10	-	-
Punta Arenas	15	18	-	-

It had been thought that the rapid depletion of radioactive debris from the USSR 1958 tests would result in an increased percentage contribution of U.S. Hardtack debris during the period of peak fallout in the spring of 1960. This does not appear to be the case, however, and suggests that U.S. Hardtack debris also had a fast fallout rate during 1959. The actual W^{185} concentration in the air in the Northern Hemisphere during the spring of 1960, when corrected for decay to the corresponding period in 1959, is only 20-25% of the 1959 value, which would indicate that over 3/4 of the W^{185}-containing Hardtack debris disappeared from the Northern Hemisphere reservoir in a year's time. The few calculations possible on the depletion rate of W^{185} in the Southern Hemisphere indicate that the rate is definitely less than in the Northern Hemisphere, unfortunately no comparison can be made during the period of peak Southern Hemisphere fallout because of insufficient W^{185} activity at that time to warrant isolation of this radionuclide.

Contribution of French Nuclear Tests to Atmospheric Contamination During 1960

As indicated in an earlier section, debris from the French nuclear tests of February and April 1960 were detected in a rather narrow band lying between 26 N and 2 S along the 80th meridian. Radiochemical analyses, however, did indicate the presence of trace quantities of some of the shorter-lived fission products in a much wider area (Thule, Greenland, to Antofagasta, Chile). Outside the primary area of detection the contribution of Sr^{90} from the first French test to the total present in the air during the month of March 1960 was negligible, as indicated in Table 6. However, the overall contribution of Sr^{90} from this source amounted to about 10% of the total burden of Sr^{90} in the tropospheric air along the 80th meridian during March. On the basis of fission yield comparisons alone, it is obvious that this test contributed less than 0.1% to the world-wide inventory of Sr^{90} (a fission yield of 60-70 kilotons vs a total fission yield exceeding 90 megatons). The fact that only traces of the shorter-lived activities (Ce^{141}, Sr^{89}, Y^{91}) were detected in the May collections from a few sites and none were found in any of the June collections would indicate that negligible quantities of these debris could have entered the stratosphere.

Table 6
Contribution of French Test of 13 February 1960 to Total Sr^{90} in the Air at Various Sites During March 1960

Site	Activity (dis/min/100 s.c.m)			Contribution of French Test %
	Sr^{90}*	Sr^{90} Equivalent†	Sr^{90} Total	
Thule	0.26	0.0016	0.908	0.18
Moosonee	0.56	0.0034	0.856	0.40
Washington	0.65	0.0039	0.940	0.41
Miami	7.3	0.044	1.51	2.9
Mauna Loa	21.6	0.130	1.39	9.3
Miraflores	78.0	0.470	0.858	55.0
Guayaquil	15.7	0.095	0.170	56.0
Lima	0.34	0.0021	0.115	1.8
Chacaltaya	0.26	0.0016	0.131	1.2
Antofagasta	0.30	0.0018	0.303	0.6
Santiago	0.0	0.000	0.360	0.0
Puerto Montt	0.0	0.000	0.311	0.0
Punta Arenas	0.0	0.000	0.163	0.0

* Corrected for decay to 13 Feb. 1960.
† Based on Sr^{89}/Sr^{90} ratio of 100 at time of fission.

Natural Radioactivity

At some future time, barring new releases of fission products through nuclear testing, studies of air motions using radiotracer techniques will of necessity depend on the naturally occurring radioactive constituents of the atmosphere. The 80th meridian program of radiochemical analyses of air samples offered an excellent opportunity to obtain some

much needed information on the background levels of radium-D (Pb^{210}) in the air over a wide range of latitudes. Some results of such measurements were reported earlier in this program (7) but the analyses were discontinued because of their interference with the more important W^{185} determinations.

Pb^{210}, because of its 22-year half-life and its source in the decay of radon which, in turn, is emitted over most land areas of the globe in comparable concentrations, might be expected to be a useful tracer for long-term air movements. The data collected so far in this program do indicate that at ground level the average concentrations at a given site can vary widely from month to month; there is insufficient data on hand presently to determine if these variations are annual, seasonal, or random. As seen in Table 4, there is a definite latitude dependence of Pb^{210} with the lowest concentrations in the more southerly regions. Other data, not yet reported, show the Pb^{210} concentration to be even lower at the South Pole. This trend is what would be expected for a source of radon and, consequently, Pb^{210} in areas where land is more plentiful, followed by progressive deposition of the Pb^{210} by natural processes during its migration southward.

An indication that relatively high Pb^{210} concentrations exist at high altitudes is shown by data for Mauna Loa and Chacaltaya where the Pb^{210} concentration is comparable to that at lower elevations. The quantity of Pb^{210} that would be produced locally near Hawaii might be expected to be negligible, so that the activity level encountered at Mauna Loa should correspond in general to that at 10,000 feet altitude and 20 N latitude.

It is of interest that the quantity of Pb^{210} isolated from samples during 1960 showed it to be a major contributor to the gross airborne radioactivity in many areas and indicates such measurements of gross β activity to be of doubtful value at the present time. This, again, points up the necessity for the determination of specific radionuclides by radiochemical or other techniques if any meaningful interpretation of atmospheric radioactivity concentrations is to be attempted.

DISCUSSION

Since the conclusion of large scale nuclear testing in 1958, there has been obtained unambiguous evidence of a strong seasonal variation in the rate of influx of stratospheric bomb debris into the troposphere of the Northern Hemisphere. Two such spring maxima have been documented and a third is evident from preliminary data from 1961 collections. The first clear evidence of a spring maximum in the Southern Hemisphere was obtained during 1960 and it was, as expected, roughly 6 months out of phase with that in the Northern Hemisphere. It is therefore evident that the same meteorological processes involving the seasonal variation in mixing of stratospheric and tropospheric air are operating in both hemispheres; in the past these cycles in the Southern Hemisphere had been obscured by the transequatorial migration of debris from the areas of higher concentration in the North. It might be expected that the seasonal cycles in the two areas will be more nearly comparable in magnitude as the activity levels in the two areas become more nearly equal.

In spite of the high rate of depletion of the stratospheric reservoir of fission product activity during 1959 from rapid fallout of debris from the 1958 USSR tests, a considerable quantity of activity still remains undeposited. If Northern Hemisphere concentrations of activity in the ground-level air are compared at corresponding periods of 1959 and 1960, half residence times as short as 3-5 months (January-March) or exceeding 12 months (November) can be obtained. The longer period, obtained at a time of minimum influence of any arctic debris, must be related to the removal rate of debris from the equatorial stratosphere; the shorter time is related to the fallout of arctic debris. Migration of debris across the equator in the stratosphere must cause some of the observed depletion so that the true half-residence time must exceed one year in the tropical stratosphere for the present spatial (vertical) distribution of debris. Preliminary Sr^{90} data for January-February

1961 when compared with January 1960 indicate an even longer half-residence time (30 months). This is the trend that would be expected as the arctic debris become less prominent and thus the later values should more clearly represent the true half-residence time of the tropical source.

In the Southern Hemisphere comparisons of activity levels during the successive September-November periods of peak fallout in 1959 and 1960 show an actual increase in Sr^{90} concentration at ground level which indicate an overall increase in the burden of Sr^{90} in the stratosphere over the Southern Hemisphere. Preliminary data from early 1961 indicate that this trend is continuing. The rate of southward transfer of material across the equator in the upper atmosphere, therefore, exceeded the rate of depletion of the Southern Hemisphere reservoir. Qualitatively, it may be concluded that the half-time of transequatorial mixing in the stratosphere is of roughly the same magnitude as the half-time of deposition from the stratospheric reservoir in the Tropics, namely several years.

CONCLUSIONS

During the past year there has been a considerable decrease in fission product radioactivity concentrations in the ground-level air of the Northern Hemisphere, but in the Southern Hemisphere an actual rise in the concentrations of long-lived fission products has been observed as a result of transequatorial mixing in the stratosphere. The rate of change of activity levels with time in the Northern Hemisphere has decreased markedly as the older debris above the Tropics become the dominant source of long-lived fission products.

A half-residence time of several years is indicated for debris presently located in the tropical stratosphere; the half-period of mixing across the equator is of the same magnitude.

The existence of spring peaks in fission product levels in both hemispheres, attributable solely to seasonal variations in the meteorology of the upper atmosphere, have been documented. Preliminary data from 1961 collections indicate that the expected Northern Hemisphere spring increase is underway.

The nuclear tests held by France in the Sahara produced only transient effects on the radioactivity of the air, with the bulk of the short-lived activity appearing in a rather narrow band (26 N to 2 S). The rapid depletion with time of fission products of moderate half-lives (one to two months) indicates that negligible quantities of debris from these tests could have entered the stratosphere.

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with a mean lifetime of several years for the process. However, two years after the cessation of the major nuclear test programs, differences in the fission product compositions of the two hemispheres still exist.

The relatively small amounts of radioactivity generated by the French nuclear tests in the Sahara produced only transient effects at a few sites along the 80th meridian. The interpretation of atmospheric mixing patterns from measurements of the concentrations of the longer-lived fission products in the air was not significantly affected by these fresher debris. Fission product ratios involving some of the shorter-lived fission products did document the presence of debris from the February 1960 test in a band extending from Miami, Florida, to Guayaquil, Ecuador, with the suggestion that small amounts of debris did appear in other areas.

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